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Constraints on the Grüneisen Theory

by Steven B. Segletes

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14. ABSTRACT
While the Grüneisen theory was developed from statistical theory, it is unclear which results of statistical mechanics are intrinsically part of the macroscopic Grüneisen theory and which may be dispensed with by users of the macroscopic theory. Several constraints on entropy and specific heat, arising from the Grüneisen theory, are developed herein. Not surprisingly, such constraints are compatible with the underlying statistical theory. Nonetheless, having been derived from macroscopic thermodynamic considerations, they represent constraints that must apply to any macrothermodynamic model expressing the Grüneisen function as being independent of temperature. An alternate formulation for the Grüneisen function is also presented.

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Contents

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1. Introduction

For solids under pressure, the most commonly used equation of state in analysis and computations is the Mie-Grüneisen equation of state. By employing the assumption that the Grüneisen function, Γ, is expressible as a function of volume, V, alone (*i.e.*, independent of temperature, T), the equation of state can be formulated to relate the pressure, p, and energy, E, back to conditions along a known "reference curve" at the same volume. It is given (*1*) as

$$p - p_{\text{ref}} = \frac{\Gamma}{V}\left(E - E_{\text{ref}}\right) \quad . \tag{1}$$

In equation 1, the subscript "ref" refers to values along a known reference function, evaluated at the volume of interest. Thus, with Γ, p_{ref}, and E_{ref} all being functions of volume alone, equation 1 expresses p as a function of E and V.

Equation 1 follows directly from the macroscopic thermodynamic definition of the Grüneisen function (*1*, *2*),

$$\Gamma = V\left(\frac{\partial p}{\partial E}\right)_V = \frac{V}{C_v}\left(\frac{\partial p}{\partial T}\right)_V \quad , \tag{2}$$

when operating under the assumption of temperature independence of Γ. In equation 2, C_v is the local value of constant-volume specific heat.

On the statistical scale of atomic oscillators, the Grüneisen function, under the assumption of temperature independence, may be expressed (*1*, *3*) as

$$\Gamma = -\frac{V}{\omega}\,\frac{d\omega}{dV} \quad , \tag{3}$$

where ω is the characteristic vibrational frequency of the atomic lattice, a function of volume only. The original assumption of Grüneisen was that the component vibrational frequencies, ω_i, of each atomic vibrational oscillator obeyed this relationship to yield identical values of Γ_i for each oscillator in the ensemble.

The Mie-Grüneisen equation of state was derived with statistical mechanics as a starting point (*1*). And yet, it is not immediately clear which results of statistical mechanics are

intrinsically *built in* to the Grüneisen theory and which may be dispensed with by one who limits himself to a macroscopic application of equation 1, with the underlying definition of equation 3.

A number of thermodynamic constraints (*4–7*) have already been demonstrated for Grüneisen materials, which reflect the coupled nature of the shock-Hugoniot, the Grüneisen function, the cold-pressure curve, *etc.* This report will add several new constraints to that list.

In this report, the equations that characterize isentropic transitions are employed to study the response of the Mie-Grüneisen equation of state (equation 1). We find that certain underlying facets of statistical theory are fundamentally, yet tacitly, carried along as part of the Grüneisen theory. In essence, it will be shown that the use of equation 1 poses constraints on the functional behaviors of entropy and specific heat.

2. Theory

In many computational settings of the Mie-Grüneisen equation of state, the reference curve is taken as the principal Hugoniot (*i.e.*, states achieved by shock compression), presumably since most high-pressure data has been gathered that way. In that case, the reference energy (subscript H for Hugoniot) is known (*1, 2*) through the Rankine Hugoniot relation as

$$E_H - E_0 = (p_0 + p_H)(V_0 - V)/2 \quad , \tag{4}$$

where subscript 0 denotes the initial "preshocked" state.

Another popular choice for the reference curve is the cold curve (*i.e.*, the 0° isotherm). In this case, the reference pressure (subscript C) is known from the isentropic relation that governs along the cold-curve:

$$p_c = -\frac{dE_c}{dV} \quad . \tag{5}$$

In materials for which $\Gamma = \Gamma(V)$, we now examine the behavior of temperature and internal energy as a function of the characteristic lattice frequency for the case of thermodynamic transitions which occur along an isentrope.

2.1 Isentropic Temperature *vs.* Characteristic Frequency

Slater (*3*) presents the thermodynamic relation

$$\mathsf{T} dS = C_v d\mathsf{T} + \mathsf{T}\left(\frac{\partial p}{\partial \mathsf{T}}\right)_V dV \quad , \tag{6}$$

where S is the entropy. However, at constant volume, as is the condition under which equation 2 operates, equation 6 becomes

$$\mathsf{T}(\partial S)_V = C_v(\partial \mathsf{T})_V \quad . \tag{7}$$

In light of this, equation 2 may be expressed as

$$\Gamma = \frac{V}{\mathsf{T}}\left(\frac{\partial p}{\partial S}\right)_V \quad . \tag{8}$$

However, the thermodynamic identity (*3*)

$$\left(\frac{\partial p}{\partial S}\right)_V = -\left(\frac{\partial \mathsf{T}}{\partial V}\right)_S \tag{9}$$

may be directly substituted into equation 8 to obtain

$$\Gamma = -\frac{V}{\mathsf{T}}\left(\frac{\partial \mathsf{T}}{\partial V}\right)_S \quad . \tag{10}$$

To the author's knowledge, this potentially useful form for the Grüneisen function is not prevalent in the literature. The function could be directly measured if, for example, the instantaneous temperature of an adiabatically expanding material could be directly measured (along with its instantaneous volume). A uniaxial-strain "plate-slap" setup could prove essential in this regard. However, in addition to the difficulty of measuring temperature itself, the need to separate the heat of plastic work from the temperature of isentropic compression could prove challenging for compressions above the Hugoniot elastic limit (HEL).

A comparison of equation 10 to equation 3 reveals that

$$\left(\frac{\partial \mathsf{T}}{\mathsf{T}}\right)_S = \frac{d\omega}{\omega} \quad . \tag{11}$$

When integrated along an isentrope, the solution in this case is that the isentropic temperature must remain proportional to the characteristic frequency,

$$(T)_S \propto \omega \quad . \tag{12}$$

2.2 Isentropic Energy *vs.* Characteristic Frequency

Consider now the Mie-Grüneisen equation of state with the 0° cold curve as the reference such that $p_{\text{ref}} = p_c$ and $E_{\text{ref}} = E_c$.

In light of equation 5, equation 1 becomes

$$p + \frac{dE_c}{dV} = \frac{\Gamma}{V}(E - E_c) \quad . \tag{13}$$

The pressure may be defined in terms of the volume-derivative of energy at constant entropy, $p = -(\partial E/\partial V)_S$. And since the cold-curve reference is also an isentrope, equation 13 becomes

$$\frac{V}{\Gamma}\frac{\partial}{\partial V}(E - E_c)_S + (E - E_c) = 0 \quad . \tag{14}$$

The quantity $(E - E_c)$ is merely the thermal component of the internal energy (at constant volume). For convenience, designate it E_{th}. The equation of state may therefore be expressed as

$$\frac{V}{\Gamma}\frac{\partial}{\partial V}(E_{\text{th}})_S + E_{\text{th}} = 0 \quad . \tag{15}$$

Along an isentrope, this homogeneous partial differential equation becomes ordinary. Its solution has been obtained (*8*, *9*) through equation 3 as

$$(E_{\text{th}})_S \propto \omega \quad . \tag{16}$$

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3. Result

In the previous section, two useful relations were established for materials obeying the Grüneisen theory—namely, equations 12 and 16 , where the temperature and thermal energy along an isentrope are proportional to the characteristic frequency of the lattice vibration. It is now shown how those relations lead to two thermodynamic constraints

upon Grüneisen materials. While the implications may not be thermodynamically surprising, they nonetheless represent real constraints upon any thermodynamic model purporting to satisfy the assumption of $\Gamma = \Gamma(V)$.

3.1 Entropy

Equation 12 indicates that temperature remains proportional to the characteristic lattice frequency during isentropic transitions. Thus, that ratio of characteristic frequency to temperature, which is fixed along an isentrope, can actually be used to identify the particular isentrope corresponding to the ratio. That is to say, entropy may be functionally characterized in terms of this ratio:

$$S = S(\omega/T) \quad . \tag{17}$$

That Grüneisen theory implies that the validity of equation 17 is compatible with the classical theory of statistical mechanics. For a system of oscillators, statistical theory indeed expresses the entropy as a function of ω/T (*3*):

$$S = k \sum_j \left(-\ln\left(1 - \exp\left[-\frac{h\omega_j}{kT}\right]\right) + \frac{\frac{h\omega_j}{kT}}{\exp\left[-\frac{h\omega_j}{kT}\right] - 1} \right) \quad , \tag{18}$$

where h is Planck's constant, k is Boltzmann's constant, and the summation is taken over the ensemble of atomic oscillators. And while the Grüneisen theory doesn't imply the functional form of this latter statistical result, it still remains constrained by equation 17 such that *the entropy must remain a function of* ω/T. The quantity $h\omega/k$ is a frequent grouping in statistical mechanics and is denoted as the characteristic temperature, Θ.

3.2 Specific Heat

Consider now the thermal energy, E_{th}. Definitionally, it can be defined as the integral of specific heat at constant volume,

$$E_{th}(V,T) = \left(\int_0^T C_V d\tau \right)_V \quad . \tag{19}$$

In the most general case, one must assume that C_V is a function not only of temperature (over which the integrand is integrated), but also a function of the particular fixed volume at which the integration takes place. We seek to determine if the Mie-Grüneisen

equation of state intrinsically leads to a thermodynamic constraint which limits the generality of the $C_V = C_V(V, T)$ assumption.

Substituting this definition into equation 16 and taking the constant of proportionality to be $h\widehat{C}/k$, one obtains

$$\frac{\left[\left(\int_0^T C_V d\tau\right)_V\right]_S}{h\omega/k} = \left(\widehat{C}\right)_S \quad . \tag{20}$$

The grouping $h\omega/k$ may be substituted with the characteristic temperature, Θ. However, ω and Θ are functions of volume only. And since the integral of specific heat is taken at constant volume, the term Θ may be moved into the integral to give

$$\left[\left(\int_0^{T/\Theta} C_V \, d(\tau/\Theta)\right)_V\right]_{(T/\Theta)} = \left(\widehat{C}\right)_{(T/\Theta)} \quad . \tag{21}$$

Since equation 17 states that an isentrope can be defined by its value of ω/T (and thus T/Θ), the isentropic constraint on both sides of equation 21 has been replaced with the constraint of fixing the value of T/Θ.

In equation 21, the limits of integration for all points on a given isentrope remain fixed, regardless of V. However, equation 21 is valid for all isentropes (*i.e.*, for all values of T/Θ), not just one. Therefore, equation 21 may be generalized as

$$\left(\int_0^{T/\Theta} C_V \, d(\tau/\Theta)\right)_V = \widehat{C} \quad , \tag{22}$$

where $\widehat{C}$ becomes a function of T/Θ to reflect the different values of proportionality (*i.e.*, $E_{th}/[h\omega/k]$) for the different isentropes. The only manner in which equation 22 can be generally satisfied at different volumes is if C_V itself is also a function of T/Θ alone so that the integral becomes independent of the volume, V, at which the integration is taken.

This constraining result arises purely from the assumption of $\Gamma = \Gamma(V)$, leading to equations 12 and 16. It is compatible with the methods of statistical mechanics, which lead to actual expressions for C_V as functions of Θ/T (the inverse of T/Θ), such as the methods of Einstein and/or Debye (*3*). So, as in the case of entropy, an equation of state assuming $\Gamma = \Gamma(V)$ does not, of itself, dictate the functional relation of the specific heat, C_V. However, *the specific heat is nonetheless constrained to remain solely a function of* T/Θ.

4. Conclusion

It is reassuring to find that the assumption of temperature independence of the characteristic vibrational frequency produces thermodynamic constraints which are compatible with statistical mechanics. Given the statistical-mechanics origins of the Mie-Grüneisen equation of state, this result is not surprising. Nonetheless, these constraints have been derived from purely macrothermodynamic arguments exclusive of statistical considerations.

It has been shown that materials obeying the assumption of $\Gamma = \Gamma(V)$ are constrained to express entropy and specific heat as functions of ω/T (or, alternately, of T/Θ). Exact functional relationships are not, however, dictated by the Grüneisen theory.

In light of the underlying statistical mechanics, such a result may seem natural. However, the current result, in fact, shows that functional restrictions on entropy and specific heat must be viewed as a *constraint* on the Grüneisen theory itself and not merely as a *natural approach* which draws from the statistical-mechanics theory. Any macrothermodynamic model which purports to describe a material with a temperature independent Grüneisen function is thermodynamically constrained to simultaneously describe entropy and C_V as sole functions of ω/T (or, alternately, of T/Θ).

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